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1. Untranslatable words are replaced with asterisks (\*\*\*).
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## CLAIM + DETAILED DESCRIPTION

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### [Claim(s)]

[Claim 1] The manufacture method of the amino alkoxysilane characterized by making magnesium, halogenated hydrocarbon, and secondary amine react at the first process, generating a magnesium amide compound and making this magnesium amide compound and alkoxysilane react at a dibasic process.

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### [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the new manufacture method of an amino alkoxysilane compound.

[0002]

[Description of the Prior Art] Many amino silane compounds which have Si-N binding are proposed as a catalyst component for raising the stereoregularity of polymer in an alpha olefin polymerization. For example, JP,3-74393,A A number, 7-118320, 7-173212, In 8-100019 each gazette, they are annular amino group content silane compounds, such as a piperidyl machine and a pyrrolidyl machine, to alkyl (hydrocarbon amino) dialkoxy silane or JP,H8-120021,A, and 8-143621 each gazette. The polymerization method of the alpha olefin to be used is indicated.

[0003] N-Si binding and RO-Si As the manufacture method of an amino alkoxysilane compound of having both bindings, (1) The alkoxy halogenation silane compound and the 2nd A reaction with a class amine compound, [ which are classified into a dehydrochlorination reaction ] (2) A reaction with the metal salt of the alkoxy halogenation silane compound or alkoxysilane compound classified into a demetal halogenation reaction or a demetal alkoxide reaction, and a secondary amine compound is mentioned.

[0004] (1) although many reactions which boil, attach and are performed under existence of a

hydrogen chloride supplement agent are known (Industrial and Engineering [ ] -- [ Chemistry and ] 1947, the 39th volume, 1368 pages, Organosilicon Compounds, Part 1, 1965, and 76-82 A page, Academic Press Inc. and big multi-annular par hydronalium amino group in three dimensions There is no example applied to manufacture of the silane compound which it has. Moreover, although the alkoxy halogenation silane compound of a raw material is obtained from a halogenation silane compound and an alcohol, manufacture of an alkoxy halogenation silane compound with purity high with disproportionation is not easy.

[0005] (2) it is alike, set and, generally manufacture the metal salt of a secondary amine compound at the reaction of the organometallic compound of an alkali metal or an alkaline earth metal, and a secondary amine compound. The reaction of the lithium amide and alkyl (bird alkoxy) Silang which are obtained at the reaction of an organolithium compound and amine is JP,3-74393,A. It is indicated in the number gazette. The synthetic method of the diamino dimethoxysilane which reacts tetramethoxy silane to JP,H7-224902,A and 8-143621 each gazette with the amino magnesium halide obtained at the reaction of organic magnesium halide and amine is indicated concretely. (2) Although the alkoxysilane compound as a raw material which can be obtained easily can be used in \*\*\*\*\*, let the expensive organometallic compound which handling takes cautions be a raw material. Especially as for an organolithium compound, an intense decomposition reaction occurs by contact with air and moisture difficultly [ synthesis of itself ]. the reaction of former metallic amide and an alkoxysilane compound -- an amino group -- three-dimensional -- dramatically -- \*\* -- there is no report of the synthetic process of the amino alkoxysilane compound which it moreover has by a high multi-annular par hydronalium amino group.

[0006]

[Problem(s) to be Solved by the Invention] The method of manufacturing the amino alkoxysilane compound which was excellent as a polymerization catalyst component, especially JI (multi-annular amino) dialkoxy silane in a high yield and an easy process is offered.

[0007]

[Means for Solving the Problem] This invention makes magnesium, halogenated hydrocarbon, and secondary amine react at the first process, generates a magnesium amide compound, and relates to the manufacture method of the amino alkoxysilane characterized by making this magnesium amide compound and an alkoxysilane compound react at a dibasic process.

[0008]

[Embodiment of the Invention] [ the manufacture method of the amino alkoxysilane of this invention ] It is characterized by making magnesium, halogenated hydrocarbon, and secondary amine react at the first process, generating a magnesium amide compound and making this magnesium amide compound and an alkoxysilane compound react at a dibasic process.

[0009] halogenated hydrocarbon of this invention -- it can be and a methyl group, an ethyl group, a propyl group, an isopropyl group, butyl, isobutyl, a hexyl group, an amyl group, an octyl group, a phenyl group, etc. can be mentioned as an example of a hydrocarbon group. Chlorine, bromine, and iodine are desirable by a halogen atom.

[0010] it is the organic amine compound with which the secondary amine compound was usually known in this invention -- especially -- many -- annular -- the 2nd Class amine and multi-annular par hydronalium 2nd A class amine compound is used suitably. Multi-annular par hydronalium 2nd as an example of a class amine compound Par hydronalium indole, par hydronalium isoindole, par hydronalium quinoline, Par hydronalium isoquinoline, par hydronalium carbazole, par hydronalium imino stilbene, Par hydronalium acridine and par hydronalium benzo[f] Quinoline and par hydronalium benzo[g] Quinoline, Par hydronalium benzo[g] Isoquinoline, the amine compound which a cyclohexyl ring like par hydronalium phenanthridine condensed, Some hydrogen atoms furthermore combined with the carbon atom in these amine compounds can mention the amine compound replaced with the alkyl group, the phenyl group, and the cycloalkyl machine. A multi-annular par hydronalium secondary amine compound has the isomer of cis- corporeal one and a transformer object in itself. In JI (multi-annular par hydronalium amino) dialkoxo silane, therefore, JI (Sis multi-annular par hydronalium amino) dialkoxo silane, (Sis multi-annular par hydronalium amino) There are three kinds of isomers of dialkoxo silane and JI (transformer multi-annular par hydronalium amino) dialkoxo silane (transformer multi-annular par hydronalium amino). [0011] As a desirable multi-annular par hydronalium secondary amine compound, par hydronalium indole, par hydronalium isoindole, par hydronalium quinoline, par hydronalium isoquinoline and those displacement derivatives, Sis, and transisomer can be mentioned especially. Although amines have absorptivity, water content is usually 1. Below weight % is 0.1 preferably. Below weight % is 0.03 weight % especially preferably. It is the following. Generally, since amines oxidize easily under existence of oxygen and it becomes the cause of coloring, before using it for a reaction, as for dissolved oxygen, being replaced and deoxidized with inert gas, such as nitrogen, is desirable.

[0012] In this invention, in the first process, the reaction of magnesium, halogenated hydrocarbon, and a secondary amine compound is performed, and a magnesium amide compound is generated. The polarity or the inert hydrocarbon solvent which does not have active hydrogen performs this reaction, and it is usually performed as for example, a polar hydrocarbon solvent in an ether solvent or the ether solvent which contains an inert hydrocarbon solvent in part. As an ether solvent, cyclic ether, such as dialkyl ether, such as diethylether, diisopropyl ether, dibutyl ether, and JIISO amyl ether, tetrahydrofuran, tetrahydropyran, and dioxane, etc. is mentioned. As an inert hydrocarbon solvent, pentane, hexane, heptane, octane, cyclohexane, mineral oil, benzene, toluene, xylene, etc. are

mentioned. Especially desirable things are low-boiling point hydrocarbon solvents, such as pentane, hexane, and heptane.

[0013] Although the magnesium metal in particular used in the first process is not restricted in this invention, since the surface has oxidized during preservation, what washes with a hydrogen chloride aqueous solution and removed surface magnesium oxide, for example is desirable. As for the addition order of a reaction, the method of adding the mixture of halogenated hydrocarbon and an amine compound and the method of adding halogenated hydrocarbon in slurry of magnesium and an amine compound are usually mentioned into slurry of magnesium.

[0014] The amount of each component used is not restricted in particular, although it is usually 1/1/1 in the molar ratio of magnesium / halogenated hydrocarbon / amine. reaction temperature -- usually -20 from -- 150 \*\* -- it is 100 \*\* from 10 preferably, and is 20 to 80 degrees C especially preferably. It is desirable to contact a little three components with high reaction temperature until a reaction begins, for after a reaction start to add a reaction component gradually, to contact, and to continue a reaction for high \*\*\*\*\*. A reaction can be easily started by adding a small amount of magnesium amide compounds beforehand prepared at the time of a reaction start, absolute alcohols, or solid iodine. Reaction time is usually 5 to 12 hours especially preferably in 3 preferably from 1 for 20 hours for 30 hours. When the reaction temperature in the first process is too high and the deposit solid of 2 magnesium halides is looked at in part by the solution, or when unreacted magnesium remains, it can also dissociate and remove before the reaction of the following process.

[0015] Although it is also the feature of this invention, the reaction solution which contains a magnesium amide compound while 2 magnesium halides or an unreacted magnesium metal had lived together can be used for the reaction of the following process as it is. Moreover, although organic magnesium halide is manufactured previously and amino magnesium halide is conventionally manufactured at the reaction with amine after that, when manufacturing organic magnesium halide, the side reaction accompanied by generation of 2 magnesium halides occurs easily. However, according to this invention, in order to manufacture amino magnesium halide, without manufacturing organic magnesium halide and isolating, generation of 2 magnesium halides by a side reaction can be suppressed.

[0016] At a dibasic process, the magnesium amide compound and alkoxysilane compound which were generated at the first process are made to react, and amino alkoxysilane is manufactured. As an alkoxysilane compound, tetra-alkoxysilane, alkyltrialkoxysilane, Dialkyl dialkoxysilane, halogeno trialkoxysilane, dihalogeno dialkoxysilane, Trihalogeno alkoxysilane, alkyl halogeno dialkoxysilane, alkyl dihalogeno alkoxysilane, dialkyl halogeno alkoxysilane, those mixtures, etc. are mentioned, and it is 1 to 24 as a carbon number of an alkyl group and an alkoxy group. Among this, they are most suitably used by tetra-alkoxysilane and

alkyltrialkoxysilane, and as an example Tetramethoxy silane, tetra-ethoxy silane, tetra-isopropoxysilane, methyl trimetoxysilane, ethyltrimethoxysilane, propyltrimethoxysilane, butyltrimethoxysilane, hexyl trimethoxysilane, etc. are mentioned. Especially tetramethoxy silane is desirable.

[0017] At this reaction, a magnesium alkoxy halogenated compound generates in addition to a desired resultant, therefore this magnesium alkoxy halogenated compound is detached by filtration or centrifugality as an insoluble solid product, and desired amino alkoxy silane can be isolated from the remaining reaction solutions by distillation etc.

[0018] in the reaction of a dibasic process -- contact of each component -- usually -20 - 140 °C -- desirable -- 0-100 °C -- 20 to 80 degrees C and time can be especially performed from 3 preferably for 10 hours for 1 to 15 hours. The amount of each component used of a magnesium amide compound and an alkoxy silane compound changes by the molar ratio of an amide group and the amino alkoxy silane which is an object. In the case of diamino dialkoxy silane, the molar ratio of a magnesium amide compound / alkoxy silane compound is usually 1-5 preferably 0.5 to 20 times. It is twice. In the case of alkylamino dialkoxy silane, the molar ratio of a magnesium amide compound / alkyltrialkoxysilane compound is usually 0.5-3 preferably 0.1 to 10 times. It is twice. In order to shorten reaction time, it is desirable to enlarge a molar ratio not much, but when loss of an unreacted magnesium amide compound is large and a magnesium amide compound is expensive, a cost of production becomes high. In order to avoid loss of an expensive magnesium amide compound, it is desirable to make a molar ratio small, but if it is made not much small, when diamino dialkoxy silane is required, it will carry out subraw [ of the amino trialkoxysilane ]. Although the contact order in particular of each component is not limited, it is desirable to add only the quantity of a request of desired alkoxy silane to a magnesium amide compound.

[0019] [ what can be suitably manufactured as amino alkoxy silane of a product ] in this invention Are JI (multi-annular par hydronalium amide) dimethoxysilane and alkyl (multi-annular par hydronalium amide) dimethoxysilane, and as the example JI (par hydronalium iso KINORINO) dimethoxysilane, JI (par hydronalium KINORINO) dimethoxysilane, Ethyl (par hydronalium iso KINORINO) dimethoxysilane, propyl (par hydronalium iso KINORINO) dimethoxysilane, [ butyl (par hydronalium iso KINORINO) dimethoxysilane is mentioned, and ] when it is JI (par hydronalium iso KINORINO) dimethoxysilane Three kinds of isomers, JI (cis-par hydronalium ISOKINORINO) dimethoxysilane, JI (tolan spur hydronalium ISOKINORINO) dimethoxysilane, and dimethoxysilane (cis- (tolan spur hydronalium ISOKINORINO) par hydronalium ISOKINORINO), are mentioned.

[0020] The amino alkoxy silane of this invention can be used as a polymerization catalyst component of alpha olefin.

[0021]

[Example] A work example is given to below and it explains still more concretely about this invention. This invention is not limited only to these work examples.

[0022] Work-example 1 thermometer, a flowing-back condenser tube, and a dropping funnel After putting in stirrer piece in a flask with a capacity of 1000ml which it had and carrying out nitrogen displacement of the inside of a flask using a vacuum pump, In 150ml, 50mg of iodine, and a dropping funnel, it is [ metal / magnesium / tetrahydrofuran / 4.85g and ] 150ml about 21ml and tetrahydrofuran in par hydronalium isoquinoline (transformer/cis- = 1/3.2) 30ml (0.2mol) and n-butyl chloride. It put in. It is a mixed solution gradually from a dropping funnel, agitating the inside of a flask One fourth It was dropped, and the water-bath temperature of the flask was controlled, reaction temperature was raised to 40 degrees C, and the reaction was started. Dropping was ended over about 5 hours at 40 more degrees C, it is at 55 degrees C and the reaction was continued for further 5 hours. Then, filtration separation of the residue was carried out and the reaction solution was obtained. Next, 29.8ml of tetramethoxy silane was again put into the dropping funnel, and it kept at 60 degrees C, and was dropped in 3 hours. Then, it is n, distilling off tetrahydrofuran at 70 to 80 degrees C. An equivalent amount of heptane was added and the reaction was continued for 8 hours. During this period and tetrahydrofuran are 200ml. A degree distills off, and it divides into 2 times, and is 200ml. n Heptane was added to the flask. Filtration separation of the magnesium metal of a raw material and the solid of the methoxy magnesium chloride generated at the reaction is carried out, it washes 3 times by heptane 30ml, and filtrate and washing liquid are distilled. Although it is an object, it is JI (par hydronalium iso quinoly) dimethoxysilane. (transformer transformer and transformer Sis and \*\*\*\*- cis isomer are about 6/36/58) It obtained. Purity [ in / boiling points are 181 \*\*/1mmHg, and / gas chromatography ] is 97.5%. It is and trimethoxysilane and par hydronalium isoquinoline were detected as an impurity (par hydronalium iso KINORINO). The yield in the raw material par hydronalium isoquinoline basis of JI (par hydronalium iso KINORINO) dimethoxysilane at this time was 77.6%.

[0023] They are cis- 50 mol % and transformer 50 mol % as work-example 2 par hydronalium isoquinoline. It reacted like the work example 1 except having used the thing. As a result, it turned out that the isomer of Torrance Torrance, Sis Torrance, and Sis Sis is detected in order in gas chromatography, and the obtained JI (par hydronalium iso quinoly) dimethoxysilane is about 25%, 50%, and 25% of mixture respectively. Purity is 97.1%. It is and trimethoxysilane and par hydronalium isoquinoline were detected as an impurity (par hydronalium iso KINORINO). The yield in the raw material par hydronalium isoquinoline basis of JI (par hydronalium iso KINORINO) dimethoxysilane at this time was 78.9%.

[0024] Work-example 3 thermometer, a flowing-back condenser tube, and a dropping funnel After putting in stirrer piece in a flask with a capacity of 1000ml which it had and carrying out nitrogen displacement of the inside of a flask using a vacuum pump, 4.85g and isopropyl ether

for a magnesium metal 150ml, In 50mg of iodine, and a dropping funnel, it is 150ml about 21ml and tetrahydrofuran in par hydronalium isoquinoline (transformer/cis- = 1/3.2) 30ml (0.2mol) and n-butyl chloride. It put in. It is a mixed solution gradually from a dropping funnel, agitating the inside of a flask One fourth The reaction was started, as it was dropped, the water-bath temperature of the flask was controlled and reaction temperature was kept at 55 degrees C. Dropping was ended over about 5 hours at 55 degrees C, it is at 60 degrees C and the reaction was continued for further 5 hours. Next, 29.8ml of tetramethoxy silane was again put into the dropping funnel, and it kept at 60 degrees C, and was dropped in 3 hours. Then, it is n, distilling off tetrahydrofuran at 70 to 80 degrees C. An equivalent amount of heptane was added and the reaction was continued for 8 hours. During this period and tetrahydrofuran are 200ml. A degree distills off, and it divides into 2 times, and is 200ml. n Heptane was added to the flask. Filtration separation of the magnesium metal of a raw material and the solid of the methoxy magnesium chloride generated at the reaction is carried out, it washes 3 times by heptane 30ml, and filtrate and washing liquid are distilled. Although it is an object, it is JI (par hydronalium iso quinoly) dimethoxysilane. (transformer transformer and transformer Sis and \*\*\*\*- cis isomer are about 6/36/58) It obtained. Purity [ in / boiling points are 181 \*\*/1mmHg, and / gas chromatography ] is 97.0%. It is and trimethoxysilane and par hydronalium isoquinoline were detected as an impurity (par hydronalium iso KINORINO). The yield in the raw material par hydronalium isoquinoline basis of JI (par hydronalium iso KINORINO) dimethoxysilane at this time was 85.4%.

[0025] They are cis- 50 mol % and transformer 50 mol % as work-example 4 par hydronalium isoquinoline. It is a work example 3 except having used the thing. It reacted similarly. As a result, it turned out that the isomer of Torrance Torrance, Sis Torrance, and Sis Sis is detected in order in gas chromatography, and the obtained JI (par hydronalium iso quinoly) dimethoxysilane is about 25%, 50%, and 25% of mixture respectively. Purity is 96.8%. It is and trimethoxysilane and par hydronalium isoquinoline were detected as an impurity (par hydronalium iso KINORINO). The yield in the raw material par hydronalium isoquinoline basis of JI (par hydronalium iso KINORINO) dimethoxysilane at this time was 86.2%.

[0026]

[Effect of the Invention] Acquisition can manufacture amino alkoxysilane by easy operation only from a possible raw material easily at a high yield, without using the organometallic compound which reactivity is dramatically high and handling takes cautions as a direct raw material by the manufacture method of this invention. it has N-H binding especially with low reactivity -- many -- annular -- the 2nd [ amine compound / class ] three-dimensional -- dramatically -- \*\* -- JI (multi-annular amino) dialkoxy silane, such as JI (multi-annular par hydronalium amino) dimethoxysilane which has two high multi-annular par hydronalium amino groups, can be manufactured easily -- them -- as an alpha olefin polymerization catalyst

component It is useful. Moreover, in this invention, the isomer molar fraction of a multi-annular par hydronalium amino group can be controlled and manufactured.

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[Translation done.]